

## **Fermicondensation**

an almost ideal glass transition

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FERMICONDENSATION - AN ALMOST IDEAL GLASS TRANSITION

By: Jeppe C. Dyre

## TEKSTER fra

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#### ABSTRACT

A fermisystem in a thermodynamic density of states evaporates into a classical gas at a finite temperature. The converse process, "fermicondensation", is shown to be close to an ideal glass transition (a transition to a state of zero entropy at a positive temperature). The fermicondensation phenomenon has a number of properties in common with Derrida's random energy model. The present study of the two models weakens the Adam-Gibbs hypothesis concerning a diverging average relaxation time at the ideal glass transition.

## 1. INTRODUCTION

Most liquids are able to form glasses when cooled sufficiently fast to avoid crystallization [1,2]. Examples of glass forming liquids include polymeric liquids, ionic liquids, molten salts or metals, simple organic liquids, and the various oxides forming ordinary glass. The glass transition is not an ordinary phase transition, but a kinetic phenomenon. The transition is not sharp and its characteristic temperature,  $T_g$ , depends somewhat on the cooling rate. There are, however, speculations that the experimental glass transition is a manifestation of an underlying genuine second order phase transition to a state of zero entropy, an "ideal glass transition". This idea was proposed by Gibbs and DiMarzio in 1958 [3]. It resolves the so-called Kauzmann paradox [4,5], i. e., the fact that <sup>the</sup> excess entropy of the supercooled liquid upon cooling extrapolates to zero at a positive temperature  $T_0$ . The excess entropy is the entropy of the liquid in excess of the purely vibrational contribution (estimated by the entropy of the crystal at the same temperature). This quantity represents the configurational entropy of the liquid and must be positive. Thus, either the extrapolation is incorrect or there is a phase transition at  $T_0$  to a state of zero configurational entropy, an "ideal glass". The real glass transition intervenes before  $T_0$  is reached, though, and the existence of an underlying ideal glass transition remains a conjecture.

Henceforth attention is focussed on the thermodynamics of the configurational degrees of freedom only; the vibrational degrees of freedom are ignored since they undergo little change at the

glass transition. Any thermodynamic system is characterized by the entropy as function of energy,  $S(E)$  [6]. The temperature is defined by

$$\frac{1}{T} = \frac{\partial S}{\partial E} \quad (1)$$

The requirements of a positive temperature and of a non-negative specific heat lead to

$$\frac{\partial S}{\partial E} > 0, \quad \frac{\partial^2 S}{\partial E^2} \leq 0. \quad (2)$$

For an ordinary system, the state of zero entropy is reached only at  $T=0$ . Thus, if  $E=0$  is the ground state energy, one has  $\frac{\partial S}{\partial E} \rightarrow \infty$  for  $E \rightarrow 0$ . For a system exhibiting an ideal glass transition, by definition,  $\frac{\partial S}{\partial E}$  is finite as  $E \rightarrow 0$ . These two possibilities are illustrated in Fig. 1. The ideal glass transition of Fig. 1b takes place at  $T=T_0$  where

$$\frac{1}{T_0} = \lim_{E \rightarrow 0} \frac{\partial S}{\partial E} \quad (3)$$

Gibbs and DiMarzio [3] originally argued for the existence of an ideal glass transition in polymeric liquids from a study of a lattice model. Several years later it was shown by Gujrali and Goldstein [7] that the mean field approximations made by Gibbs and DiMarzio are incorrect and that, in fact, the lattice model has a positive entropy at any nonzero temperature. But despite this negative result, the Gibbs-DiMarzio idea remains attractive since it offers a simple resolution of the Kauzmann paradox.

The standard example of a system exhibiting an ideal glass transition is Derrida's random energy model (REM) from 1980 [8] (sec. 2). It seems that few other examples exist. This paper studies the "fermi condensation" phenomenon (sec. 3), which has a number of properties in common with the REM. Both models exhibit an ideal glass transition only in a certain limit. The two models also have similar relaxation behavior, as shown in sec. 4.

## 2. THE RANDOM ENERGY MODEL

This section briefly reviews the random energy model. To motivate this model, expand  $S(E)$  to second order in  $E$  for a system exhibiting an ideal glass transition (where again it is assumed that the ground state is  $E=0$ ):

$$S(E) = \alpha_1 E - \frac{1}{2} \alpha_2 E^2 + \dots \quad (4)$$

The function  $S(E)$  is the logarithm of the density of states  $n(E)$  [6]. Thus, ignoring the higher order terms in Eq. (4) corresponds to assuming a gaussian density of states,

$$n(E) \propto \exp \left[ \alpha_1 E - \frac{1}{2} \alpha_2 E^2 \right] \quad (5)$$

The random energy model is now defined simply by choosing  $N$

energy levels randomly from the gaussian [8]. These  $N$  energies completely define the system. It is important that  $N$  is finite, only after the thermodynamics is worked out is the  $N \rightarrow \infty$  limit taken. In letting  $N$  go to infinity care must be taken to assure that the lowest energy remains close to zero; this is done by varying  $\alpha_1$  appropriately with  $N$  [8]. It is easy to see that the resulting model exhibits an ideal glass transition: Close to  $E=0$  the density of states is approximately exponential,

$$n(E) \propto \exp[\alpha_1 E], \quad (E \approx 0). \quad (6)$$

For  $T > T_0$  where  $T_0 = 1/\alpha_1$ , the maximum of the canonical probability,  $n(E) \exp(-E/k_B T)$ , increases with increasing  $T$ . The average energy,  $\bar{E}(T)$ , is an increasing function of temperature so the specific heat, and thereby the entropy, is positive. Below  $T_0$ , on the other hand, the Boltzmann factor dominates over  $n(E)$ , driving the system to the lowest energy state. Thus, the entropy is zero. Actually, the entropy is only strictly zero in the  $N \rightarrow \infty$  limit, but even for a finite system there is a sharp decrease in entropy below  $T_0$ .

### 3. FERMICONDENSATION

A system of fermions is usually thought of in terms of occupied and unoccupied states, where it is implied that the

majority of the particles are to be found in occupied states. For some fermisystems, however, this picture is adequate only at low temperatures whereas at higher temperatures most particles are classical, i. e. , have energies for which the average occupation number is  $\ll 1$  . Consider a system of non-interacting fermions in a "thermodynamic" density of states  $g(\epsilon)$  , i. e. , where  $S(\epsilon) = \ln(g(\epsilon))$  obeys Eq. (2). Expanding  $S(\epsilon)$  around the zero temperature chemical potential (the fermi energy),  $\mu_0$  , leads to

$$g(\epsilon) = g_0 \exp \left[ c_1 (\epsilon - \mu_0) - \frac{1}{2} c_2 (\epsilon - \mu_0)^2 + \dots \right] . \quad (7)$$

At low temperatures most particles have energies close to  $\mu_0$ . But at higher temperatures the Boltzmann tail of the fermifunction is not able to suppress the  $\exp[c_1(\epsilon - \mu_0)]$  part of  $g(\epsilon)$  . The result is that most particles move to higher energies. The particles become classical in the sense that, for the majority of particles, the average occupation number is much less than one. The transition from a fermi to a classical system is continuous and not a sharp transition. It takes place around  $T=T_0$  where  $T_0$  is given by

$$T_0 = \frac{1}{k_B c_1} . \quad (8)$$

The transition upon cooling from high temperatures may be termed a "fermicondensation", since here the fermisea is formed from the classical gas. The term "gas" is relevant for the high temperature state where the particles effectively do not "interact" via the fermi exclusion principle.



The fermion condensation is signalled by a decrease in specific heat below  $T_0$ . In Fig. 2 the specific heat per particle is shown for two different values of  $\lambda = c_1/\sqrt{c_2}$  for a system with a gaussian density of states (Eq. (7)). Above  $T_0$  most of the fermions have "evaporated". The particles move independently and each particle has a specific heat of  $1/(2k_B T^2 c_2)$ , as is easy to show. Below  $T_0$  the specific heat per particle is small as in an ordinary fermion system. For large  $\lambda$  the transition is close to a phase transition. For  $\lambda \rightarrow \infty$  an ideal glass transition takes place since in this limit the specific heat, and thereby the entropy, vanishes below  $T_0$ .

We now briefly discuss relaxation in the model. Assume the simplest possible realistic dynamics, namely that where the jump rate of a fermion depends only on the energy of the initial state [9]. This corresponds to the case where the fermion, once it is excited into the transition state, has forgotten where it came from and may end up in any other (unoccupied) state. If the energy of the transition state is  $\epsilon_0$ , the relaxation time for jumps from a state of energy  $\epsilon$  is

$$\tau = \tau_0 e^{\frac{\epsilon_0 - \epsilon}{k_B T}} \quad (\epsilon < \epsilon_0) \quad (9)$$

where  $\tau_0$  is a microscopic time. A cut-off at  $\epsilon_0$  has been introduced to ensure that no state has larger energy than the transition state. Now, below  $T_0$  most particles have energy close to  $\mu_0$  since the density of states is exponentially increasing. Thus, relaxation is Arrhenius with an activation energy given by

$$\Delta \varepsilon = \varepsilon_0 - \mu_0 \quad (10)$$

Above  $T_0$ , the average energy of the particles,  $\bar{\varepsilon}$ , depends on temperature. Since the particles here move independently,  $\bar{\varepsilon}(T)$  is determined by maximizing the probability for one particle having energy  $\varepsilon$ :

$$\left. \frac{d}{d\varepsilon} \left[ g(\varepsilon) e^{-\frac{\varepsilon}{k_B T}} \right] \right|_{\varepsilon = \bar{\varepsilon}(T)} = 0. \quad (11)$$

Because  $g(\varepsilon)$  is by assumption a thermodynamic density of states,  $\bar{\varepsilon}(T)$  is an increasing function of temperature so the activation energy of  $\tau$ ,

$$\Delta \varepsilon = \varepsilon_0 - \bar{\varepsilon}(T), \quad (12)$$

decreases with increasing temperature.

Figure 3 shows a so-called Arrhenius plot of  $\tau$ , i. e.,  $\ln(\tau)$  as function of  $T^{-1}$ . The behavior of Fig. 3 is like that seen in experiments on the real glass transition. Above the glass transition one observes an average relaxation time which is more than Arrhenius temperature dependent, while in the glassy phase  $\tau$  is Arrhenius [10]. In experiment, there is furthermore a sudden decrease in the slope of the Arrhenius plot at  $T_g$ , just like in Fig. 3. However, one should not attach too much significance to the similarity between fermioncondensation and experiments on the real liquid-glass transition. The real glass transition is a kinetic phenomenon and, for instance, the activation energy of in the glassy phase depends on the cooling rate at which the glass

was formed.

#### 4. DISCUSSION

Fermicondensation is an alternative to the random energy model as a model exhibiting an almost ideal glass transition. Unlike the REM, no randomness is invoked in fermicondensation. Therefore, no ensemble averaging is needed to evaluate physical properties like the specific heat, as is necessary for a more accurate treatment of the REM than given here [8]. Since both models involve a gaussian density of states and since the fermions above  $T_0$  move independently, the fermisystem behaves like the REM above  $T_0$ . In particular, if the assumption of transition rates being function of the initial energy only is made also for the REM [11], the two models have similar relaxation behavior. Note that the behavior of  $\tau$  shown in Fig. 3 is valid for the REM even below  $T_0$ : Here the REM is in its ground state so the activation energy of  $\tau$  becomes constant. Thus, the two models are similar both as regards their thermodynamics and their relaxation behavior.

Adam and Gibbs have argued that at an ideal glass transition the average relaxation time becomes infinite because of the few available states to jump into [12]. This argument is still controversial, but this study certainly weakens the Adam-Gibbs hypothesis. In both models relaxation in the glassy phase takes place with a finite relaxation time which is, as a matter of fact, smaller than expected by extrapolating the behavior above  $T_0$ .

To summarize, the fermicondensation phenomenon provides an abstract model for the ideal glass transition. No randomness is invoked in the model, but otherwise the model has a number of features in common with the random energy model. The present study of the two models does not confirm the Adams-Gibbs hypothesis concerning a diverging average relaxation time at  $T_0$ .

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## FIGURE CAPTIONS

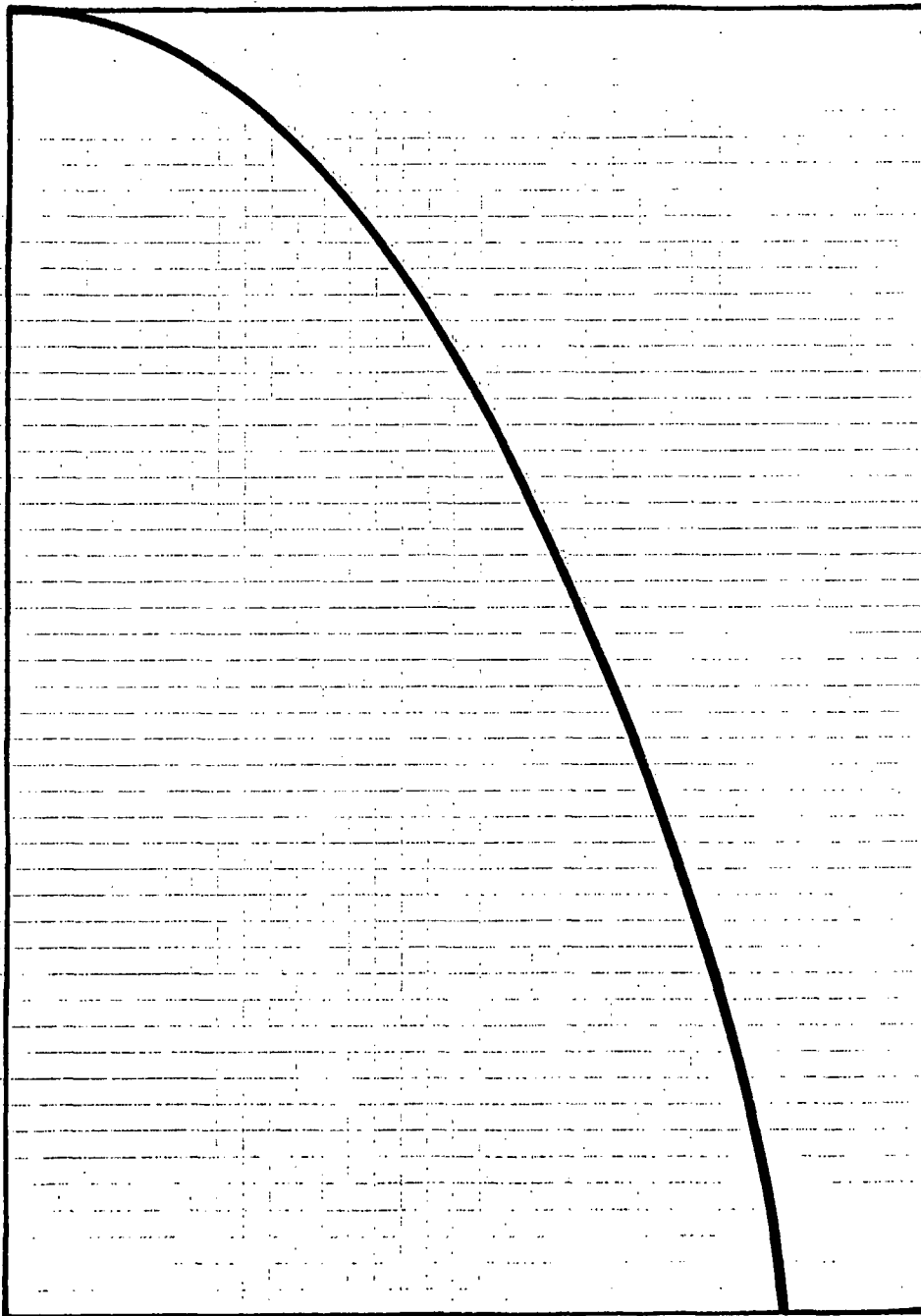
Fig. 1: Entropy as function of energy for an ordinary system (Fig. 1a) and for a system exhibiting an ideal glass transition (Fig. 1b). Temperature is defined by  $\frac{1}{T} = \frac{\partial S}{\partial E}$  [6] and thermodynamics requires  $S(E)$  to be an increasing function of energy with a non-positive second derivative. If  $S(E)$  has a finite slope at  $E=0$ , there is a transition to a state of zero entropy at a positive temperature (Fig. 1b). This is the ideal glass transition, which is a second order phase transition.

Fig. 2: Specific heat per particle as function of temperature for a fixed number of fermions in a gaussian density of states (Eq. (7)). The two curves show the case of (a)  $\lambda = 8$  and (b)  $\lambda = 20$  where  $\lambda = c_1/\sqrt{c_2}$ . The specific heat is given in units of  $k_B c_1^2/2c_2$ . Around  $T=T_0$  where  $T_0$  is given by Eq. (8) the fermisea "evaporates". Above  $T_0$  the fermions behave as independent classical particles, each with a specific heat  $\propto T^{-2}$ . Upon cooling, the classical gas "condenses" into the fermisea, the fermicondensation.

Fig. 3: Logarithm of the average relaxation time  $\tau$  plotted as function of inverse temperature in the fermicondensation model where  $\tau$  is given by Eq. (9) (schematic drawing). Above  $T_0$   $\tau$  is more than Arrhenius temperature dependent while below  $T_0$   $\tau$  becomes Arrhenius, much like for the real laboratory glass transition. The activation energy of  $\tau$  is the slope of the secant

marked by dots (and not the slope of the tangent marked by the punctuated line, as is often assumed). Upon cooling the activation energy of  $\tau$  increases until  $T_0$  is reached. The activation energy then becomes constant. As discussed briefly in sec. 4, the random energy model has a similar relaxation behavior.

$S(E)$



$E$

Fig. 1a



$S(E)$

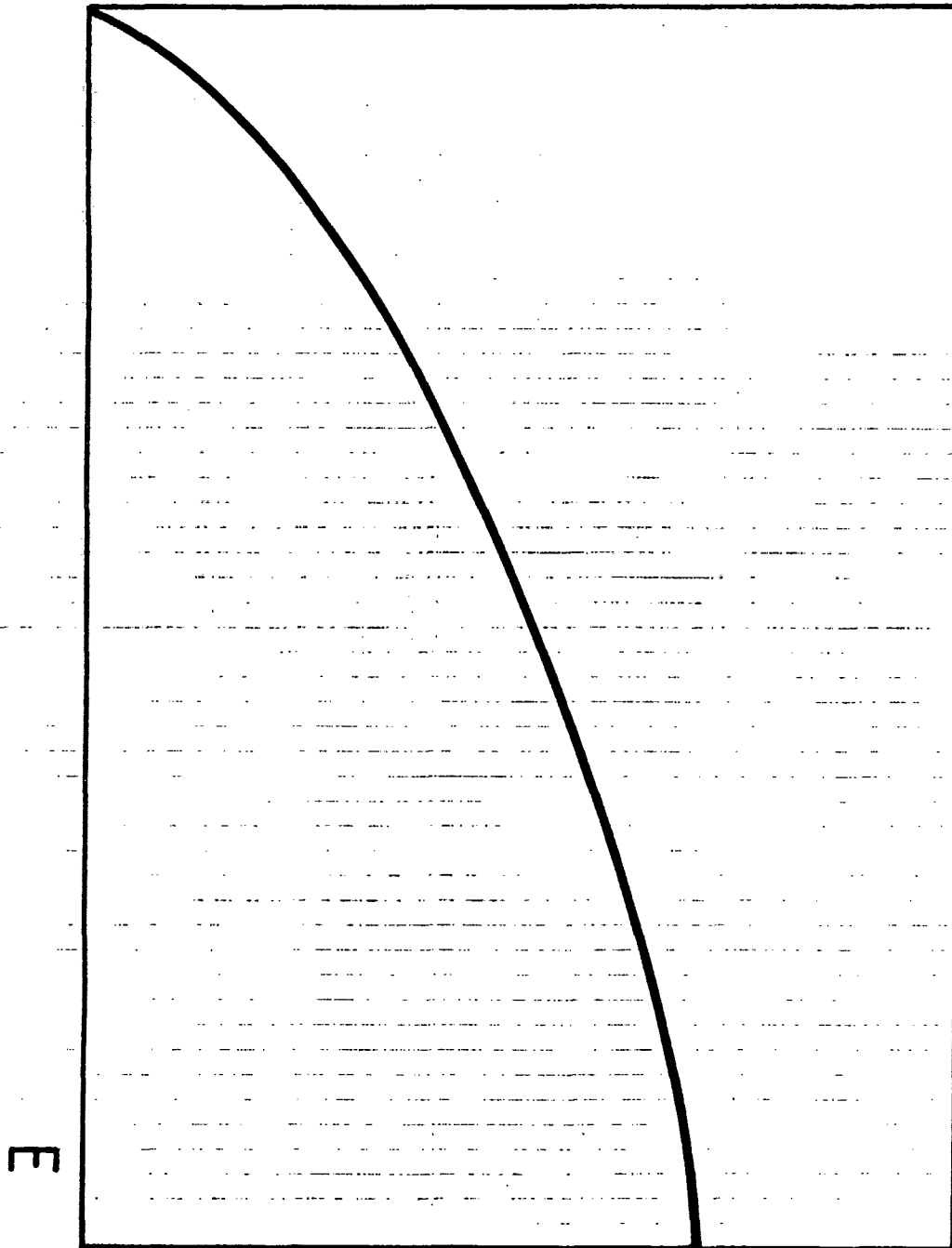


Fig. 1b

Fig. 2

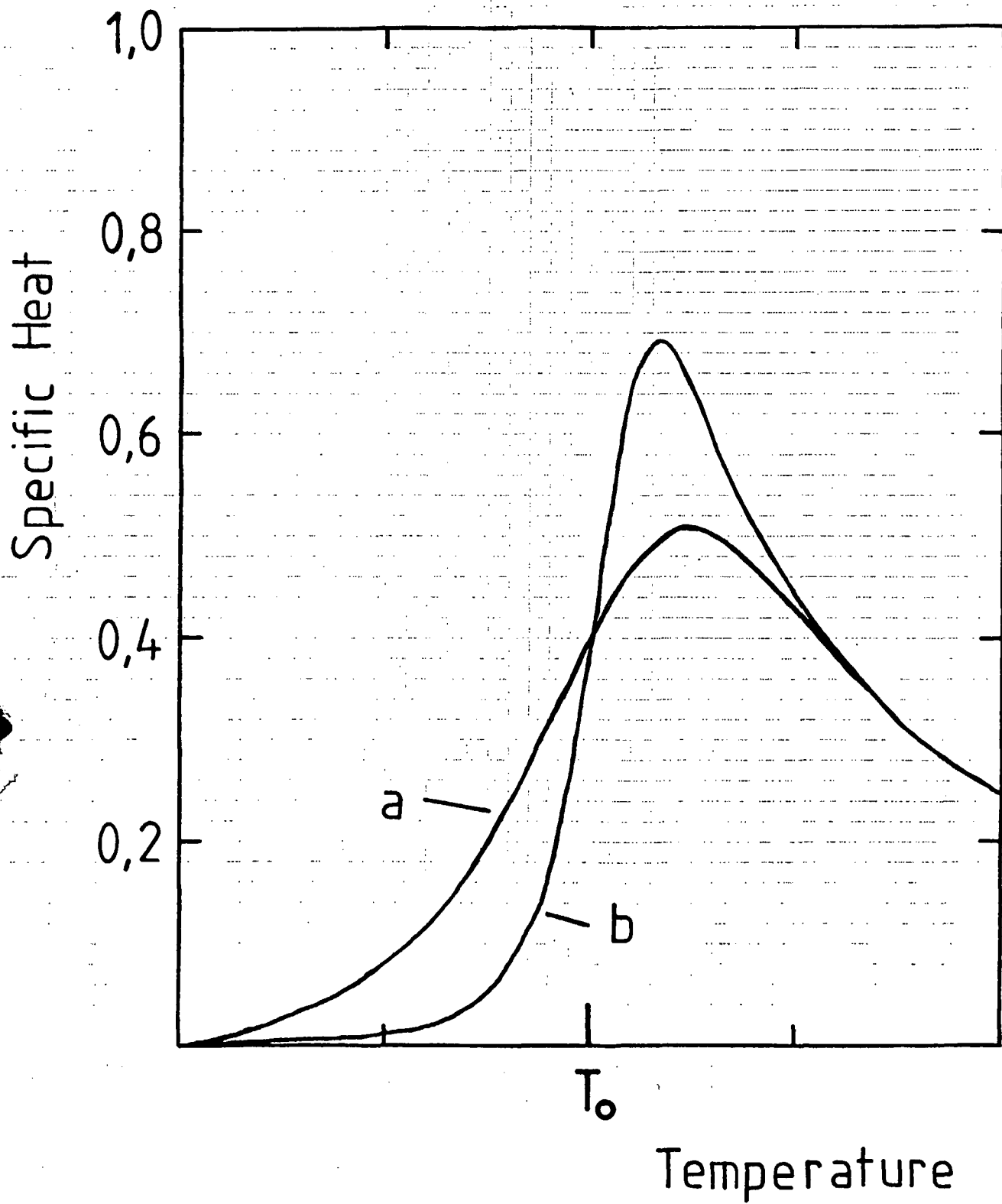
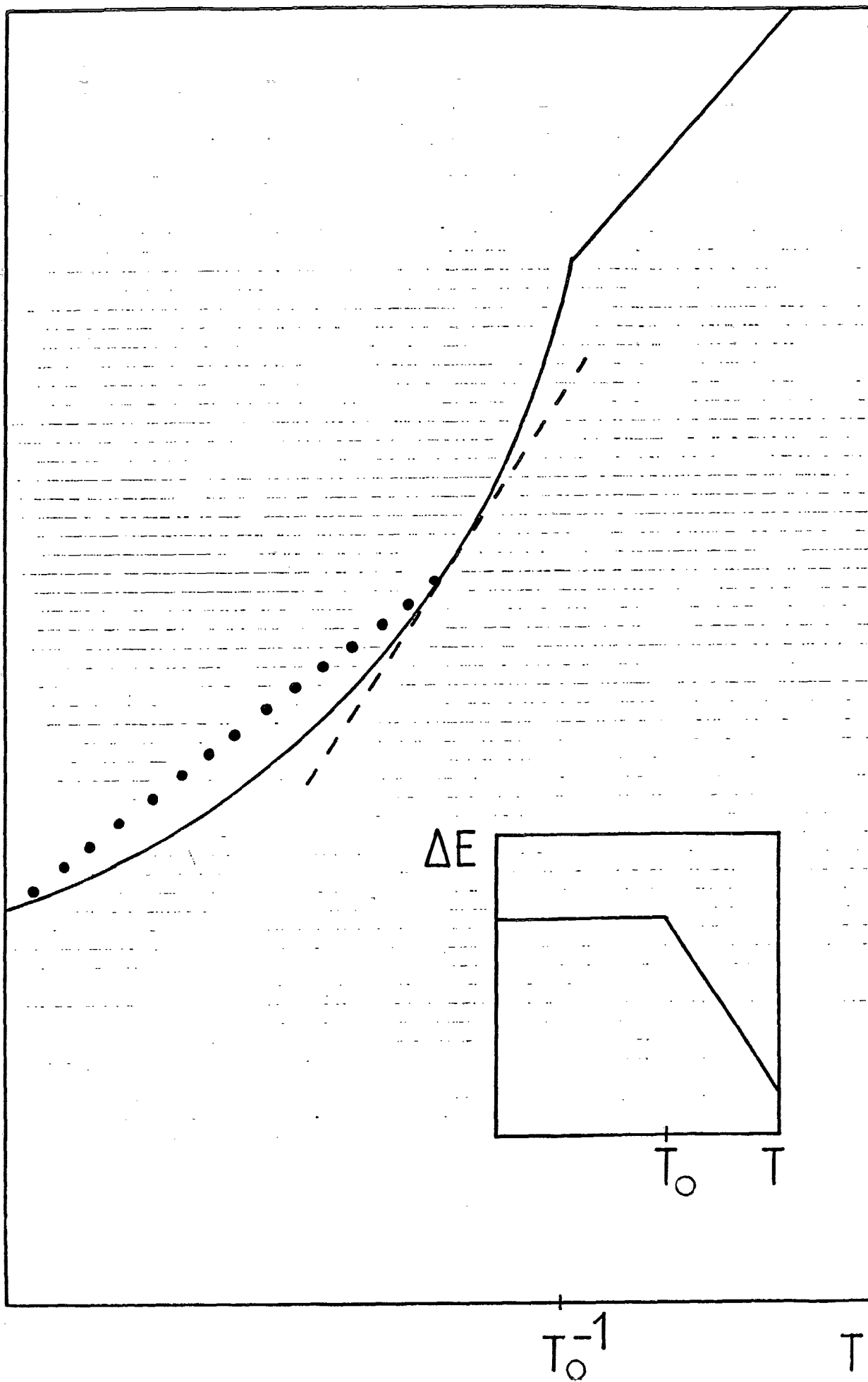


Fig 3

Fig.3

 $\ln(\tau)$ 

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Vejleder: Peder Voetmann Christiansen.  
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Projektrapport af Finn C. Physant  
Vejleder: Ib Thiersen

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Krzysztof P. Wojciechowski

143/87 "Kursusmateriale til Matematik på NAT-BAS"

af: Mogens Bruh Heefelt

144/87 "Context and Non-Locality - A Peircan Approach"

Paper presented at the Symposium on the Foundations of Modern Physics The Copenhagen Interpretation 60 Years after the Como Lecture. Joensuu, Finland, 6 - 8 august 1987.

By: Peder Voetmann Christiansen

145/87 "AIMS AND SCOPE OF APPLICATIONS AND MODELLING IN MATHEMATICS CURRICULA"

Manuscript of a plenary lecture delivered at ICMTA 3, Kassel, FRG 8.-11.9.1987

By: Mogens Niss

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- en ny frekvensbaseret målemetode.

Fysikspeciale af Jan Vedde

Vejledere: Niels Boye Olsen & Petr Višćor

147/87 "Rapport om BIS på NAT-BAS"

redigeret af: Mogens Bruh Heefelt

148/87 "Naturvidenskabsundervisning med Samfundsperspektiv"

af: Peter Colding-Jørgensen DLH  
Albert Chr. Paulsen

149/87 "In-Situ Measurements of the density of amorphous germanium prepared in ultra high vacuum"

by: Petr Višćor

150/87 "Structure and the Existence of the first sharp diffraction peak in amorphous germanium prepared in UHV and measured in-situ"

by: Petr Višćor

151/87 "DYNAMISK PROGRAMMERING"

Matematikprojekt af:  
Birgit Andresen, Keld Nielsen og Jimmy Staal  
Vejleder: Mogens Niss

152/87 "PSEUDO-DIFFERENTIAL PROJECTIONS AND THE TOPOLOGY OF CERTAIN SPACES OF ELLIPTIC BOUNDARY VALUE PROBLEMS"

by: Bernhelm Booss-Bavnbek  
Krzysztof P. Wojciechowski

153/88 "HALVLEDERTEKNOLOGIENS UDVIKLING MELLEM MILITÆRE OG CIVILE KRÆFTER"

Et eksempel på humanistisk teknologihistorie  
Historiespeciale

Af: Hans Hedal

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Dirichlet feedback control problems"

by: Michael Pedersen

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AF: Karin Beyer, Sussanne Blegaa, Birthe Olsen,  
Jette Reich, Mette Vedelsby

163/88 "EN MATEMATISK MODEL TIL BESTEMMELSE AF PERMEABILITETEN FOR BLOD-NETHINDE-BARRIEREN"

Af: Finn Langberg, Michael Jarden, Lars Frellesen  
Vejleder: Jesper Larsen

164/88 "Vurdering af matematisk teknologi Technology Assessment Technikfolgenabschätzung"

Af: Bernhelm Booss-Bavnbek, Glen Pate med  
Martin Bohle-Carbonell og Jens Højgaard Jensen

165/88 "COMPLEX STRUCTURES IN THE NASH-MOSER CATEGORY"

by: Jens Gravesen



166/88 "Grundbegreber i Sandsynligheds-  
regningen"

Af: Jørgen Larsen

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Af: Jørgen Larsen

167b/88 "BASISSTATISTIK 2. Kontinuerte  
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undersøgt ved Mössbauerspektroskopi.

Fysikspeciale af:

Birger Lundgren

Vejledere: Jens Martin Knudsen  
Fys.Lab./HCØ

169/88 "CHARLES S. PEIRCE: MURSTEN OG MØRTEL  
TIL EN METAFYSIK."

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1891-93.

Introduktion og oversættelse:

Peder Voetmann Christensen

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Kvantemekanikkens grundlagsproblem  
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Fysikprojekt af:

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af: Morten Andersen, Ulla Engström,  
Thomas Gravesen, Nanna Lund, Pia  
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om kaos"

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MOGENS NISS, Roskilde (Denmark)

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og Ole Møller Nielsen

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188/90 "FERMICONDENSATION - AN ALMOST IDEAL GLASS TRANSITION"

by: Jeppe Dyre